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FEASIBILITY STUDY OF RARE EARTH SEMICONDUCTOR LASERS OF THE TYP--ETC(U)
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The possibility of making a rare earth or rare earth doped semiconductor laser is investigated. The material examined in detail is Y_2HfS_5, which is of the general class of materials Ln_2TX_5, where Ln is a rare earth, T is zirconuim or hafnium, and X is sulfur or selenium. For Y_2HfS_5:Nd, the Stark split energy levels of $Nd^{(3+)}$ are determined in the energy range 0 to 2 eV (the band gap of Y_2HfS_5). The individual line to line tran- → next page

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sition probabilities are calculated assuming S_4 symmetry for the Nd^{3+} site, and the branching ratios are given. The calculated branching ratio for spontaneous emission of the $^4F_{3/2}$ to the $^4I_{11/2}$ is 0.574, and the next largest branching ratio of the $^4F_{3/2}$ to $^4I_{9/2}$ is 0.298. The branching ratio for the $^4I_{11/2}$ is larger in this material than the calculated branching ratio for Nd in YAG, which is 0.480. The transition probabilities are used to calculate the excitation of the various multiplets by conduction electrons.

CONTENTS

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TABLES

1. INTRODUCTION

In this report, the possibility is discussed of making a rare earth doped semiconductor laser. Such a laser would have the principal advantage of being pumped with conventional ac or dc circuitry, as well as other advantages such as adaptability to miniaturized circuits. The chief obstructions to the development of such a device are (1) the crystal composition that precludes the substitution of rare earths into the material or (2) the relatively narrow band gap of most semiconductors that results in the absorption of the laser radiation once the rare earth is substituted.

Recently developed semiconductor materials of the general type Ln_2TX_5 do not have these difficulties, where Ln is yttrium or one of the triply ionized lanthanides, T is zirconium or hafnium, and X is sulfur or selenium. In particular, Y_2HfS_5 has been reported[1] to have an approximate band gap of 2 eV, which is more than adequate to allow passage of the 1.3 eV $^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser radiation normally observed in Nd^{3+}. Furthermore, the doping of Nd^{3+} into the isovalent Y^{3+} site may be achieved to any degree desirable.

On these bases, there is a good possibility that $Nd:Y_2HfS_5$ will be a successful semiconductor laser material. There are other factors, however, that must be considered. For example, there must be a sufficient number of conduction electrons to pump[2,3] the rare earth ions, but few enough so that the mean free paths of laser photons are long enough to span a large number of excited ions. Secondly, the density of the rare earth ions must be low enough so that self-quenching is not severe, yet high enough to produce sufficiently intense radiation. It is known that self-quenching, for example, depends quite sensitively on the type of host material, but, at present, the determination of this host dependence seems possible only by experiment.

Many of these questions regarding the performance of the proposed material $Nd:Y_2HfS_5$ can be answered only experimentally. However, to examine its feasibility from a theoretical point of view, the reported crystal structure[4] of Y_2HfS_5 has been used to perform a lattice sum and to determine theoretical Stark splittings of Nd^{3+} at the Y^{3+} site.

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[1] *P. C. Donohue and W. Jeitschko, Mat. Res Bull., 9 (1974), 1333.*

[2] *D. C. Krupka and D. M. Mahoney, J. Appl. Phys., 43 (1972), 2314.*

[3] *Hiroshi Kobayashi, Shosaku Tanaka, Hiroshi Sasakura, and Yoshihiro Hamakawa, Japanese J. Appl. Phys., 13 (1974), 1110.*

[4] *W. Jeitschko and P. C. Donohue, Acta Cryst., B31 (1975), 1890.*

These results are given in section 2. In section 3, the calculations are discussed that determine which Nd levels are populated by conduction electrons as a function of the applied electric field. A summary of the results is given in section 4, where it is concluded that there is a good possibility of pumping Nd^{3+} impurity ions by conduction electrons in Y_2HfS_5, and laser action should therefore be possible.

2. CRYSTAL FIELD CALCULATION

A first estimate of the crystal field parameters, B_{nm}, in the Hamiltonian

$$H_x = \sum_{n,m} B_{nm}^{+} \sum_i C_{nm}(\hat{r}_i) \qquad (1)$$

for the rare earth ions in a crystal may be obtained by performing a lattice sum over the host ions.[5] To perform the lattice sum, it is necessary to know the position of each atom in the unit cell and the type of crystalline structure. These data have been reported by Jeitschko and Donohue[4] for Y_2HfS_5 and are given in table I. The point symmetry of the yttrium site (which will be occupied by substitutional rare earths) is C_1 and would allow, in general, all the A_{nm} (lattice sums) for n even, which are proportional to the B_{nm}. As is shown in table II, all the A_{nm} are nonzero so that for *even* n the total number of parameters for f electrons is 27. We have chosen to reduce this large number of parameters by approximating the symmetry to a higher symmetry for calculational purposes. In obtaining this approximation, we have made use of the rotational invariants, A_n, defined by

$$A_n = \left[\frac{1}{2n+1} \sum_m |A_{nm}|^2\right]^{1/2}, \qquad (2)$$

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[4] *W. Jeitschko and P. C. Donohue, Acta Cryst., B31 (1975), 1890.*

[5] *Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare Earth Ion-Host Interactions, 4. Intensity Calculations and Derived B_{km} for the Lanthanides, Harry Diamond Laboratories TR-1807 (1977).*

where the A_{nm} are the lattice sums. We let each approximated lattice sum parameter be

$$A'_{nm} = \eta_n A_{nm}(x) \tag{3}$$

and require the new rotational invariant to be identical to the value in equation (2), which was obtained by using the true lattice sums A_{nm}. The $A_{nm}(x)$ are the lattice sum parameters obeying the higher symmetry. In the cases chosen, they are $A_{nm}(C_s)$ for $m = 0, 2, 4, \leq n$ and $A_{nm}(S_4)$ for $|m| = 0, 4, \leq n$, n *even* and $|m| = 2, 6$, n *odd*. The lattice sums along with the approximate values obtained for these two symmetries are shown in table II. The even-fold crystal field parameters, B_{nm}, are them obtained by assuming that[5]

$$B_{nm} = \rho_n A'_{nm} , \tag{4}$$

where for Nd^{3+}, $\rho_2 = 0.1706$, $\rho_4 = 0.5776$, and $\rho_6 = 1.5897$, and these values were used to obtain B_{nm} for both C_s and S_4 symmetry. The low lying energy levels of the crystal split spectra are given in table III for both C_s and S_4. As can be seen, the S_4 approximation does not alter the overall splitting of most of the multiplets and appears to be reasonably good for many of the individual levels. The B_{nm} (n *even)* along with the A'_{nm} (n *odd)* were used to calculate the electric dipole matrix elements, M_{ij}, between the various levels in S_4 symmetry. These matrix elements will be used in the calculation of the excitation of Nd^{3+} by hot electrons (carriers).

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[5]*Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare Earth Ion-Host Interactions, 4. Intensity Calculations and Derived* B_{km} *for the Lanthanides, Harry Diamond Laboratories TR-1807 (1977).*

TABLE I. CRYSTALLOGRAPHIC DATA AND ATOMIC POSITIONS FOR Y_2HfS_5*

Orthorhombic space group P_{nma}†

Four molecules per unit cell (Z = 4)

a = 11.4585(3) Å

b = 7.7215(3) Å

c = 7.2207(2) Å

Atom	Position	x	y	z
Y	8(d)	0.1778	0.9974	0.0251
Hf	4(m)	0.0060	¼	0.5742
S(1)	8(d)	0.4081	0.0367	0.1630
S(2)	4(m)	0.1822	¼	0.3331
S(3)	4(m)	0.5032	¼	0.5522
S(4)	4(m)	0.2921	¼	0.8125

*W. Jeitschko and P. C. Donohue, Acta Cryst., B31 (1975), 1890.

†International Tables, I, Kynoch Press, Birmingham, England (1952).

TABLE II. LATTICE SUMS FOR Y_2HfS_5 EVALUATED AT THE YTTRIUM SITE (C_1 SYMMETRY) WITH C_s AND S_4 APPROXIMATIONS

n	m	C_1		$C_s(C_{1h})$		S_4	
		Real	Imaginary	Real	Imaginary	Real	Imaginary
		A_{nm}	A_{nm}	A_{nm}	A_{nm}	A_{nm}	A_{nm}
2	0	2816	0	3223	0	4190	0
2	1	1322	573.3	0	0	0	0
2	2	1613	-370.5	1846	-424.0	0	0
4	0	-820.5	0	-913.6	0	-1135	0
4	1	146.0	190.0	0	0	0	0
4	2	-1070	43.93	-1191	48.91	0	0
4	3	851.8	4.427	0	0	0	0
4	4	-1292	330.7	-1439	368.2	-1786	457.3
6	0	-18.48	0	-25.34	0	-36.07	0
6	1	103.6	27.01	0	0	0	0
6	2	67.79	27.62	92.96	37.88	0	0
6	3	-28.75	-10.18	0	0	0	0
6	4	74.57	55.22	102.3	75.72	145.6	107.8
6	5	44.27	36.40	0	0	0	0
6	6	50.36	33.43	69.06	45.84	0	0

Odd n A_{nm} for S_4 symmetry where

A_{32} = 921.2-295.2i

A_{52} = -727.1-4548i

A_{72} = -6.43+23.66i

A_{74} = -0.20-29.53i

Charge

q_Y = +3

q_{Hf} = +1

q_S = -1.4

Note: All A_{nm} *in units* cm^{-1} $(\text{Å})^{-n}$. *The crystal field parameters,* B_{nm}, *are given by equation (4) in the text.*

TABLE III. ENERGY LEVELS OF Nd in Y_2HfS_5 for C_s and S_4 SYMMETRY

Term	$C_s(C_{1h})$ energy (cm^{-1})	S_4 energy (cm^{-1})	S_4 energy (eV)
$^4I_{9/2}$	0	0	0
	69	2	0.0002
	98	52	0.0064
	165	200	0.0248
	287	226	0.0280
	1925	1,895	0.2350
	1947	1,917	0.2377
	1,973	1,934	0.2398
$^4I_{11/2}$	1,990	1,948	0.2415
	2,025	2,070	0.2566
	2,142	2,074	0.2571
	3,897	3,875	0.4804
	3,914	3,883	0.4814
	3,942	3,903	0.4839
$^4I_{13/2}$	3,968	3,933	0.4876
	3,978	3,937	0.4881
	4,014	4,076	0.5054
	4,159	4,085	0.5065
	5,895	5,890	0.7303
	5,938	5,903	0.7319
	5,970	5,916	0.7335
	5,993	5,958	0.7387
$^4I_{15/2}$	6,007	5,970	0.7402
	6,047	6,007	0.7448
	6,091	6,190	0.7675
	6,306	6,206	0.7695
	11,415	11,383	1.4113
$^4F_{3/2}$	11,505	11,472	1.4224
	12,274	12,255	1.5194
$^4F_{5/2}$	12,367	12,320	1.5275
	12,402	12,371	1.5338
	12,530	12,537	1.5544
	12,608	12,539	1.5547
$^2H_{9/2}$	12,624	12,593	1.5614
	12,654	12,628	1.5657
	12,689	12,671	1.5710
	13,335	13,315	1.6509
$^4F_{7/2}$	13,420	13,367	1.6573
	13,461	13,431	1.6653
	13,503	13,481	1.6714

3. EXCITATION CALCULATION

A detailed theory of the actual mechanism of excitation of the electronic levels of an impurity ion in a solid by impact of hot electrons (carriers) in semiconductors is very complicated. However, by making various assumptions, a first approximation can be derived by using a variety of available sources of information.

An approximate distribution function for electrons in the presence of an electric field in a semiconductor has been derived by Baraff.[6] The approximate distribution function agrees quite closely to the exact numerical results in both the low and the high field regions (energy below and above the ionization threshold). For energies, W, below the ionization threshold, Baraff's distribution function is given by*

$$f(W) = W^{\alpha} e^{-bW} \quad , \tag{5}$$

where

$$\alpha = \frac{3eE\lambda}{2\left(2W_0 + eE\lambda\right)} \quad ,$$

$$b = \frac{3}{cE\lambda\left(2 + eE\lambda/W_0\right)} \quad ,$$

E is the electric field,

λ is the mean free path of the carrier,

W_0 is the low energy loss (optical phonon),

e is the proton charge.

The result given in equation (5) is the distribution function (unnormalized) for energies below ionization and is used here. The

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**The form of equation (5) given by Baraff is $m_O(W) = W^{-a} e^{-bW}$, where the energy distribution, $f(W)$, is given by $f(W) = W^{\frac{1}{2}} m_O(W)$. Thus, α and a are related by $\alpha = \frac{1}{2} - a$ (see D. C. Krupka, J. Appl. Phys. 43, 476 (1972).*

[6] G. A. Baraff, Phys. Rev., 133 (1964), A26.

distribution function above ionization also has been derived by Baraff. The number of excited levels of the impurity ion,[7] a rare earth in this case, is proportional to

$$n_{if} \propto \int_{W \geq \Delta_{if}}^{\infty} W^{\frac{1}{2}} f(W) \sigma\left(W, \Delta_{if}\right) dW \quad , \tag{6}$$

where $\Delta_{if} = W_f - W_i$ and $\sigma(W, \Delta_{if})$ is the energy cross section for excitation of the rare earth ion from the state i to the state f. If the dipole approximation is made in the Born approximation,[8] the cross section in equation (6) is given by

$$\sigma\left(W, \Delta_{ij}\right) \propto \frac{M_{ij}^2}{W} \ln \left[\frac{W^{\frac{1}{2}} + \left(W - \Delta_{ij}\right)^{\frac{1}{2}}}{W^{\frac{1}{2}} - \left(W - \Delta_{ij}\right)^{\frac{1}{2}}} \right] , \tag{7}$$

providing an appropriate directional average is made. In equation (7), the quantity M_{ij} is given by

$$\vec{M}_{ij} = \langle j | \vec{r} | i \rangle \ ,$$

and Δ_{ij} is the energy difference $W_j - W_i$ with W_i the energy of the lower energy state of the rare earth ion. The total number of rare earth ions excited to levels j from i can be obtained by using equations (5), (6), and (7) to obtain

$$N_{ij} = K M_{ij}^2 \Delta_{ij}^{A-1} I\left(\Delta_{ij}\right) \ , \tag{8}$$

where

K is a constant,

$A = \frac{1}{2} + \alpha$,

$$I\left(\Delta_{ij}\right) = \int_1^{\infty} x^A e^{-b\Delta_{ij}x} \ln \left[(x)^{\frac{1}{2}} + (x - 1)^{\frac{1}{2}} \right] dx \ .$$

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[7] *D. C. Krupka, J. Appl. Phys., 43 (1972), 476.*

[8] *L. I. Schiff, Quantum Mechanics, 3rd ed., McGraw-Hill Book Co., New York (1968), ch 9.*

In the case of Nd, the levels of interest are the $^4I_{11/2}$ and $^4F_{3/2}$, which are the usual levels for a solid state Nd^{3+} laser.[9]

In this first calculation, the initial state in equation (8) is taken as the lowest level of the crystalline Stark split multiplet $^4I_{9/2}$. The entire ground multiplet can be included by using appropriate Boltzmann factors, but such a refinement of the theory is unnecessary at the present stage of approximations. Thus, the total number of levels excited into the $^4I_{11/2}$ states is

$$N_{i,11/2} = K \sum_f N_{if} \quad , \tag{9}$$

where the sum on f contains only the crystal split levels of the $^4I_{11/2}$. In the excitation of the $^4F_{3/2}$, the higher terms are usually close enough to be tightly coupled by the lattice vibration. For this term, we consider

$$N_{i,3/2} = K \sum_f N_{if} \quad , \tag{10}$$

where the sum covers all the levels with Δ_{if} greater than the $^4F_{3/2}$ and less than the energy gap (assumed to be of the order of 2 eV).

Due to the large number of factors entering into equations (6), (7), and (8), it is difficult to obtain meaningful values for $N_{i,3/2}$ and $N_{i,11/2}$. Since we are dealing only with the $^4F_{3/2}$ and the $^4I_{11/2}$ levels, we denote N_1 and N_2 for these quantities in further discussions. These numbers, N_1 and N_2, would depend on such variables as the number of carriers per unit volume and the density of rare earth ions, as well as a large number of physical constants. Fortunately, for application to possible lasers, the quantities of interest are the number of ions excited into the $^4F_{3/2}$ levels as compared with the number excited into the $^4I_{11/2}$ levels. A ratio can be expressed as

$$R = N_2/N_1 \tag{11}$$

so that the constants are eliminated, and the condition of possible laser action being achieved is that R be very small.

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[9]*Robert J. Pressley, ed., Handbook of Lasers, The Chemical Rubber Company, Cleveland, OH (1971), Section 13.*

The constant W_0 given in equation (5), which is the low energy loss, can be estimated as the highest optical phonon frequencies in Y_2HfS_5. As stated by Jeitschko and Donohue,[4] the complex can be thought of as $Y_2^{3+}[HfS_5]^{-6}$, where the bonding to Y^{3+} is ionic, while internal bonding in the HfS_5 complex is covalent. This being the case, we would expect the higher optical phonon frequency to be of the order found in such covalent complexes[10,11] as WO_4 or VO_4; thus, W_0 should lie in the range 200 cm^{-1} to 1000 cm^{-1} (approximately 0.025 to 0.124 eV). The lower frequency optical modes due to ionic motion can be ignored, as is apparent in the results (higher W_0 require larger fields to achieve the same value of R).

The electric field, E, is the only external variable in equation (5) and enters in the combination $eE\lambda$. It is difficult to obtain a reasonable estimate of the mean free path, λ; consequently, in the computation we let $x = eE\lambda$ and give all the results in terms of this composite variable.

The squares of the matrix elements of the electric dipole operator, M_{ij}^2, as calculated by using the crystal field parameters in the S_4 approximation, were used in equation (8) and then in equations (9) and (10) to obtain N_1 and N_2, with i the lowest energy level of the Nd^{3+} ion. These results were then used to calculate R for a range of values of x. The results of these calculations are shown in figures 1 and 2 for several values of W_0. As can be seen, the higher the value in W_0, the larger the electric field must be (larger x) for R to be much smaller than unity. Thus, only the highest frequency lattice vibrations need to be considered, at least in a first calculation.

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[4] *W. Jeitschko and P. C. Donohue, Acta Cryst., B31 (1975), 1890.*
[10] *A. S. Barker, Phys. Rev., 135 (1964), A742.*
[11] *Edward D. Reed and H. Warren Moos, Phys. Rev. B, 8 (1973), 980.*

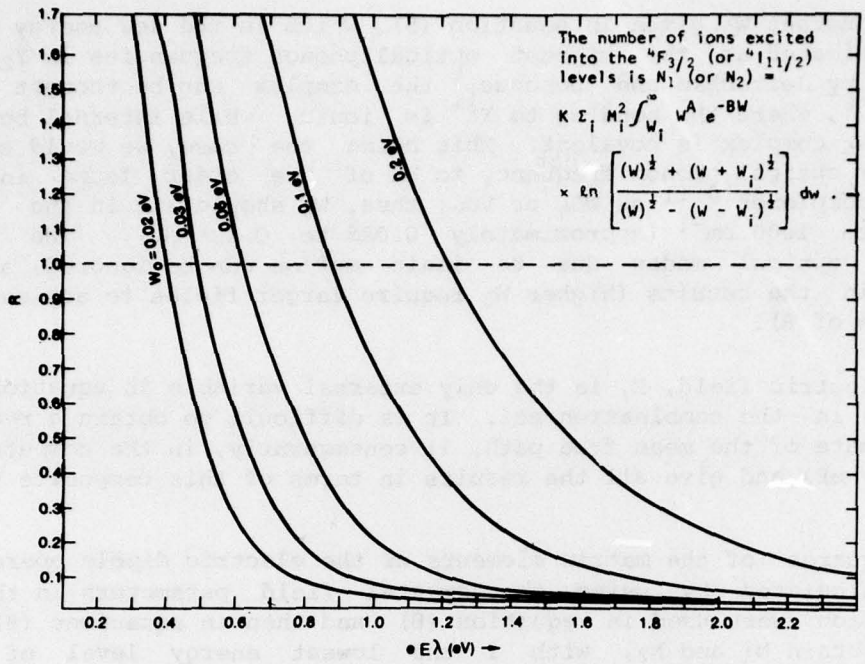

Figure 1. Ratio, R, of the number of ions, N_2, excited into the $^4I_{11/2}$ energy levels to the number of ions, N_1, excited into the $^4F_{3/2}$ levels versus the parameter eEλ.

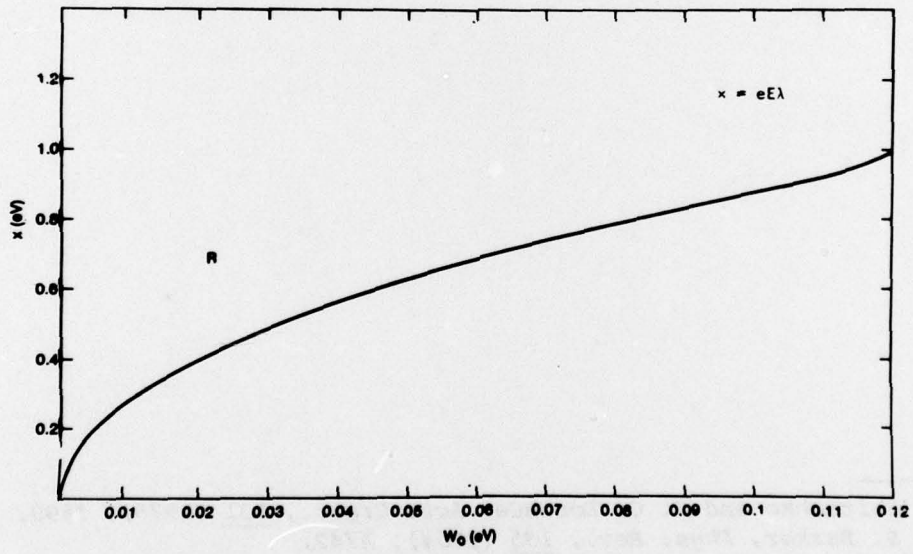

Figure 2. Values of x and W_o such that $R = N_2/N_1 = 1$.

4. DISCUSSION OF RESULTS

The primary objective of this study was to make calculations necessary to determine the feasibility of developing a new laser material, such as $Y_2HfS_5:Nd^{3+}$, that offers certain advantages over conventional gas, single crystal, or semiconductor lasers. This material is expected to (1) have a highly monochromatic output owing to a Nd^{3+} transition between $^4F_{3/2}$ and $^4I_{11/2}$ Stark split energy levels, (2) be pumped by conduction electrons, and (3) be able to be miniaturized for use in integrated optics systems.

The calculations in section 2 were made for the purpose of determining the possible output frequency of a proposed $Y_2HfS_5:Nd$ laser. By using the crystallographic data and atomic positions reported elsewhere, Nd energy levels were calculated by assuming the Nd site symmetry to be $C_s(C_{1h})$, which approximates the actual C_1 site symmetry. Since our computer programs are not yet capable of making line to line intensity calculations for such low symmetries, the Nd site symmetry was approximated by S_4. Energy levels and squared matrix elements of the electric dipole operator were then obtained.

The Stark splittings determined by assuming S_4 symmetry are not necessarily in good agreement with those obtained assuming $C_s(C_{1h})$ symmetry. The line to line intensity calculations given in table IV may likewise not be reliable. However, the branching ratios given in tables V and VI suggest that the Nd laser would operate by emission between a $^4F_{3/2}$ and a $^4I_{11/2}$ energy level at a wavelength near 1.06 μm.

As the calculations in section 3 indicate, population inversion can indeed be achieved between the $^4F_{3/2}$ and the $^4I_{11/2}$ energy levels. This inversion occurs for those values of R (R = population of $^4I_{11/2}$/ population of $^4F_{3/2}$) less than 1 as shown as a function of $eE\lambda$, where E is the applied electric field and is the only external variable. Optimum pumping conditions can be determined for a particular semiconductor such as Y_2HfS_5 by making measurements to determine λ, the mean free path of the carrier in the semiconductor.

TABLE IV. SQUARE OF MATRIX ELEMENTS, $\vec{M}_{ij} = \langle j|\vec{r}|i\rangle$, FOR $^4F_{3/2}$ MULTIPLET TO ^4I_j MULTIPLETS FOR Nd^{3+} IN Y_2HfS_5

i	W_i	$M^2_{i,27}$	$M^2_{i,28}$
1	0	9,316	1,180
2	2	7,425	1,175
3	52	587	2,685
4	200	790	14,329
5	226	5,828	15,720
6	1,895	3,784	15,796
7	1,917	3,381	2,297
8	1,934	17,150	14,470
9	1,948	22,250	7,991
10	2,070	1,606	56,620
11	2,074	6,865	42,940
12	3,875	1,197	1,020
13	3,883	977	14,669
14	3,903	1,341	294
15	3,933	1,573	16,224
16	3,937	11,042	2,395
17	4,076	1,527	12,466
18	4,085	13,442	5,652
19	5,890	507	20
20	5,903	593	935
21	5,916	95	121
22	5,958	194	704
23	5,970	791	91
24	6,007	723	876
25	6,190	1,341	652
26	6,206	3,590	64

Note: W_{27} is at 11.383 cm^{-1} and W_{28} is at 11,472 cm^{-1}. Here the S_4 symmetry approximation is used.

TABLE V. INDIVIDUAL BRANCHING RATIOS FOR LEVELS 27 AND 28 TO LOWER ENERGY LEVELS OF Nd^{3+} in Y_2HfS_5

i	W_i	Branching ratio from 27	Branching ratio from 28
1	0	0.1454	0.0093
2	2	0.1158	0.0093
3	52	0.0090	0.0209
4	200	0.0117	0.1073
5	226	0.0857	0.1169
6	1,895	0.0342	0.0725
7	1,917	0.0303	0.0105
8	1,934	0.1531	0.0656
9	1,948	0.1978	0.0361
10	2,070	0.0137	0.2460
11	2,074	0.0586	0.1863
12	3,875	0.0054	0.0023
13	3,883	0.0044	0.0335
14	3,903	0.0059	0.0007
15	3,933	0.0069	0.0363
16	3,937	0.0482	0.0054
17	4,076	0.0063	0.0264
18	4,085	0.0253	0.0119
19	5,890	0.0009	0.0000
20	5,903	0.0010	0.0008
21	5,916	0.0002	0.0001
22	5,958	0.0003	0.0006
23	5,970	0.0013	0.0001
24	6,007	0.0012	0.0007
25	6,190	0.0020	0.0005
26	6,206	0.0053	0.0000

TABLE VI. MULTIPLET BRANCHING RATIOS FOR LEVELS 27 AND 28 OF $^4F_{3/2}$ TO ^4I_j MULTIPLETS OF Nd^{3+} IN Y_2HfS_5

J	Branching ratio from 27	Branching ratio from 28	Total
9/2	0.3676	0.2636	0.2980
11/2	0.4878	0.6170	0.5743
13/2	0.1324	0.1165	0.1217
15/2	0.0122	0.0030	0.0060

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